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(54) POLYOLEFIN COMPOSITIONS HAVING IMPROVED MOLDABILITY

(71) We, IDEMITSU KOSAN KABUSHIKI KAISHA, (IDEMITSU KOSAN CO. LTD), a Japanese Body Corporate, of 1-i 3-chome, Marunouchi, Chiyoda-ku, Tokyo, Japan, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to an inorganic filler-incorporated polyolefin composition having an improved moldability.

Molding materials composed of an inorganic filler-incorporated polyolefin composition are known in the art. In such molding materials, talc, clay, calcium carbonate, calcium sulfate, calcium hydroxide, asbestos and silica are generally employed as inorganic fillers. However, these molding materials heretofore employed in the art have a poor moldability and therefore their use is limited to the preparation of boards, artificial papers and the like.

When such an inorganic filler is incorporated into a polyolefin composition, the use of a large amount of the filler is desirable in order to reduce the cost of the molded articles and to improve physical properties thereof. As the compatibility of the filler with the polyolefin is poor, however, the kneading of the polyolefin with the filler and the molding of the kneaded mixture tend to become difficult with an increase in the amount of the filler. In general, if the amount of the filler exceeds 80 parts by weight per 100 parts by weight of the polyolefin, the moldability of the resulting kneaded mixture will deteriorate considerably and if the amount of the filler exceeds 300 parts by weight per 100 parts by weight of the polyolefin, the resulting kneaded mixture will lose moldability. Accordingly, the development of a polyolefin composition which contains a large amount of an inorganic filler and which has a good moldability has been in great demand in the art.

[*Price 33p*]

It has now been found that the moldability may be improved by incorporating a small amount of a specific combination by incorporating a small amount of a specific combination of certain higher aliphatic compounds into a mixture of a polyolefin and an inorganic filler.

Accordingly the present invention provides an inorganic filler-containing, polyolefin composition, which comprises a mixture of 100 parts by weight of a polyolefin and 80 to 400 parts by weight of an inorganic filler, the said mixture having incorporated therein, a modifier which is a combination of (A) a metal salt of a higher fatty acid and (b) at least one higher C_{10} to C_{26} aliphatic compound which is a higher C_{10} to C_{26} paraffin, a higher C_{10} to C_{26} alcohol, a polyhydric alcohol ester of a higher C_{10} to C_{26} fatty acid, a higher C_{16} to C_{26} fatty acid amide, a fatty acid ester of polyethlene glycol or polybutene. When these modifier components (A) and (B) are used singly, the intended effects cannot be obtained, but when they are employed in combination, an excellent synergetic effect is attained which imparts good moldability to a polyolefin composition containing an inorganic filler.

As the higher fatty acid metal salt utilizable as the component (A) in the modifier of this invention, there can be mentioned, for example, calcium salts, zinc salts, lead salts and the like metal salts of higher fatty acids such as stearic acid, olefinic acids and lauric acid. As the higher paraffin utilizable as the component (B), there may be exemplified paraffin wax, liquid paraffin, microcrystaline wax and ethylene oligmers. Examples, of the higher alcohol include stearyl alcohol, palmityl alcohol and the like. Examples of the higher fatty acid esters with polyhydric alcohols include an ester of 1,2-dihydroxystearic acid with glycerol. Examples of the higher fatty acid amide include stearic acid amide, while examples of the fatty acid ester of polyethylene glycol include polyethylene glycol mono-

stearate. As polybutene, one having a molecular weight of 1000 to 2000 is usually employed.

Such additives may be employed in a small amount and is generally used in an amount of 0.5 to 10 parts by weight per 100 parts by weight of the total of the resin and inorganic filler.

In this invention, the components (A) and (B) of the modifier interact with each other to exhibit an enhanced lubricating effect between the polyolefin and the filler, and between the polyolefin composition and a metal die during the molding step.

Examples of the polyolefin utilizable in this invention include polyethylene, polypropylene, polybutadiene, polyisoprene, ethylene/vinyl acetate copolymers, ethylene/propylene copolymers, styrene/buradiene copolymers, polyethylene/polybutadiene blends and the like. Homopolymers, copolymers or polymer blends can be used in this invention, provided that they are composed mainly of polyolefins.

Inorganic fillers conventionally used for plastics can be used in this invention. For instance, such fillers include mineral powders composed predominantly of silicates such as talc and clay; metal salts such as calcium carbonate, calcium sulfite, calcium sulfate and calcium silicate; and metal oxides such as silica, zinc oxide and quick lime. The inorganic filler is employed in an amount of 80 to 400 parts by weight per 100 parts by weight of the polyolefin type resin.

The composition of this invention may be shaped into various forms easy to handle and may be used as blank material for molding. For instance, all of the foregoing components of the composition may be mixed and kneaded by the aid of a conventional kneading means such as an inter mixer, a Banbury mixer or the like and the resulting kneaded mixture may be pulverized by the aid of a crusher to prepare the composition in the form of small granules. Alternatively, all the components of the composition may be fed to an extruder and extruded into a rod-like form which is then cut into pellets. It is also possible to shape a kneaded mixture of all the components into a sheet by means of a mixing roll and to cut the sheet into pellets by means of an angular pelletizer.

The composition thus obtained in the form of small granules or pellets exhibits high fluidity and moldability when incorporated with a specific auxiliary component of this invention and molten by heating. The composition of this invention may easily be molded into a sheet, a rod, a pipe or other suitable form by means of a conventional molding machine such as an extruder and an injection molding machine.

When the composition of this invention is subjected to injection molding, it exhibits moldability comparable to that of conventional polyolefin compositions free of any inorganic filler. Although the moldability of such polyolefin composition generally deteriorates by incorporating thereinto an inorganic filler, such deterioration is not observed in the case of the composition of this invention. Therefore, injection molding can be performed in shortened cycles. When the composition of this invention is subjected to extrusion molding, since the composition has a good moldability, the extrusion output of the composition can greatly be increased even at a low extrusion pressure, thus enhancing the production of molded articles. Moreover, the molded articles thus prepared have a good surface condition and excellent mechanical properties.

This invention will now be illustrated in more detail by way of Example.

Example

A given amount of a polyolefin and a given amount of an inorganic filler were charged in a Banbury mixer and the mixture was kneaded for 3 minutes at a casing temperature of 150°C. Then, a given amount of a combination of (A) a higher fatty acid metal salt and (B) a higher aliphatic compound was added to the kneaded mixture and the resulting mixture was kneaded for 3 minutes.

The resulting kneaded mixture was molded into a sheet by means of a mixing roll and the sheet was cut into pellets by means of an angular pelletizer.

The resulting pellets were then molded by means of an injection molding machine. In this case, a spiral flow length was measured to evaluate the moldability. Separately, test speciments were prepared from the pellets and the tensile elasticity and tensile strength were measured to evaluate the mechanical properties. Results are shown in Table 1. The injection molding conditions adopted in the above procedures were a maximum cylinder temperature of 230°C., a pressure of 1000 kg/cm² and a metal mold temperature of 45°C. The measurement of the spiral flow length was conducted with a conventional Archimedean spiral mold. The tensile elasticity strength were measured according to ASTM D—638.

The above-mentioned pellets were molded into a sheet by means of an extruded having a die orifice diameter of 30 mm. The relation between the extrusion pressure and the extruded amount as well as the surface condition of the resulting sheet was examined to evaluate the moldability. Results are shown in Table 2. In this case, the cylinder temperature of the extruder was maintained at 200°C.

TABLE 1

Exp. No.	Composition (parts by weight)				Moldability	Mechanical Properties	
	Polyolefin	Inorganic Filler	Modifier		Spiral Flow Length (cm)	Tensile Elasticity (Kg/cm^2)	Tensile Strength (Kg/cm^2)
			Component (A)	Component (B)			
1	polypropylene [1]) (50)	$CaSO_3$ (50)	–	–	47	23,100	228
2	,,	,,	calcium stearate (0.5)	–	54	22,800	225
3	,,	,,	–	paraffin wax (2.5)	51	22,900	233
4	polyethylene [2]) (50)	$CaCO_3$ (50)	–	–	25	unmoldable	unmoldable
5	polypropylene (50)	$CaSO_3$ (50)	calcium stearate (0.5)	paraffin wax (2.5)	60	22,200	220
6	,,	,,	,,	liquid paraffin (7.5)	74	13,400	164
7	,,	talc (50)	,,	paraffin wax (2.5)	89	20,100	231
8	,,	$CaCO_3$ (50)	,,	,,	74	15,600	173
9	,,	zinc oxide (50)	,,	,,	92	12,700	201
10	,,	kaolin clay (50)	,,	,,	82	20,800	223
11	,,	$CaSO_4$ (50)	,,	,,	76	17,300	188

TABLE 1 (Continued)

Exp. No.	Composition (parts by weight)				Moldability	Mechanical Properties	
	Polyolefin	Inorganic Filler	Modifier		Spiral Flow Length (cm)	Tensile Elasticity (Kg/cm^2)	Tensile Strength (Kg/cm^2)
			Component (A)	Component (B)			
12	polypropylene (50)	$CaSO_3$ (50)	calcium stearate (0.5)	polyethylene glycol fatty acid ester[3]) (5.0)	78	20,500	188
13	,,	,,	,,	,, (7.5)	83	18,700	167
14	,,	,,	,,	higher alcohol[4]) (7.5)	87	18,800	186
15	,,	,,	,,	higher fatty acid amide[5]) (7.5)	81	22,500	183
16	,,	,,	,,	polybutene[6]) (7.5)	82	17,600	170
17	,,	talc (50)	,,	higher fatty acid polyhydric alcohol ester[7]) (7.5)	74	27,600	236
18	,,	zinc oxide (50)	,,	higher alcohol (7.5)	95	13,300	185
19	,,	$CaSO_4$ (50)	,,	polyethylene glycol fatty acid ester (7.5)	70	12,700	160

[1]) Polypropylene : tradename J 700 manufactured by Mitsui Petrochemical

[2]) Polyethylene : tradename 5100 LP manufactured by Mitsui Petrochemical

[3]) Polyethylene glycol fatty acid ester : tradename LX–3 manufactured by Kyodo Chemicals (polyethylene glycol monostearate melting at 38 – 108° C.)

[4]) Higher alcohol : tradename Karcol 86 manufactured by Kao Sekken (mixture of 85% of stearyl alcohol and 15% of palmityl alcohol melting at 57° C.)

[5]) Higher fatty acid amide : tradename VLA–1 manufactured by Kwaken Fine Chemical (melting at 126 – 130° C.)

[6]) Polybutene : tradename 300 H manufactured by Idemitsu Petrochemical (having molecular weight of 1000 – 2000)

[7]) Higher fatty acid polyhydric alcohol ester : tradename Castor Wax A manufactured by Nippon Yushi (composed mainly of 1,2-dihydroxy-stearic acid glyceride and melting above 84° C.)

TABLE 2

Exp. No.	Polyolefin type resinous composition								Extrusion pressure (Kg/cm^2)	Amount extruded (cm^3/hour)	Surface condition of sheet
	Resin		Inorganic filler		Metal salt of higher fatty acid		Higher paraffin				
	Sort	Amount [5])	Sort	Amount [5])	Sort	Amount [5])	Sort	Amount [5])			
1 [1])	poly-ethylene [2])	100	calcium carbonate	0	calcium stearate	0	paraffin wax [4])	0	95	5200	Smooth
2 [1])	,,	30	,,	70	,,	0	,,	0	130	3700	Rough
3	,,	70	,,	30	,,	0.5	,,	0.5	75	5700	Smooth
4 [1])	,,	30	,,	70	,,	0	,,	0	165	5000	White spots formed
5	,,	30	,,	70	,,	0.5	,,	7.5	67	5900	Smooth
6 [1])	poly-propylene [3])	30	calcium sulfite	70	,,	0	,,	0	175	6700	Rough
7	,,	30	,,	70	,,	0.5	,,	7.5	85	8400	Smooth

Notes

[1]) Comparative Example

[2]) Commercially available (5100 L.P, Mitsui Petrochemical Ind. Co., Ltd.)

[3]) Commercially available (J 7000, Mitsui Petrochemical Ind. Co., Ltd.)

[4]) M. P. 68 – 70°C.

[5]) Parts by weight

WHAT WE CLAIM IS:—

1. An inorganic filler-containing, polyolefin composition, which comprises a mixture of 100 parts by weight of a polyolefin and 80 to 400 parts by weight of an inorganic filler, the said mixture having incorporated therein, a modifier which is a combination of (A) a metal salt of a higher fatty acid and (B) at least one higher C_{10} to C_{20} aliphatic compound which is a higher C_{10} to C_{20} paraffin, a higher C_{10} to C_{20} alcohol, a polyhydric alcohol ester of a higher C_{10} to C_{20} fatty acid, a higher C_{10} to C_{20} fatty acid amide, a fatty acid ester of polyethylene glycol or polybutene.

2. A composition as claimed in claim 1 wherein the polyolefin is polyethylene or polypropylene.

3. A composition as claimed in claim 1 or claim 2 wherein the inorganic filler is talc, clay, calcium carbonate, calcium sulfite, calcium sulfate, calcium silicate or zinc oxide.

4. A composition as claimed in any one of the preceding wherein the higher fatty acid metal salt is a metal salt of stearic acid.

5. A composition as claimed in claim 4 wherein the metal salt of stearic acid is calcium stearate.

6. A composition as claimed in any one of the preceding claims wherein the modifier is employed in an amount of from 0.5 to 10 parts by weight per 100 parts by weight of the total of the polyolefin and inorganic filler.

7. A composition as claimed in any one of the preceding claims which is formed into granules or pellets.

8. A moulded article whenever prepared from a composition as claimed, in any one of the preceding claims.

9. A composition as claimed in claim 1 substantially as hereinbefore described with reference to any one of the experiments described in the Example.

10. A modifier for improving the moldability of an inorganic filler-containing polyolefin composition which comprises (A) a metal salt of a higher C_{10} to C_{20} fatty acid, and (B) at least one higher C_{10} to C_{20} aliphatic compound which is a higher C_{10} to C_{20} paraffin, a higher C_{10} to C_{20} alcohol, a polyhydric alcohol ester of a higher C_{10} to C_{20} fatty acid, a higher C_{10} to C_{20} fatty acid amide, or a fatty acid ester of polyethylene glycol or polybutene.

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